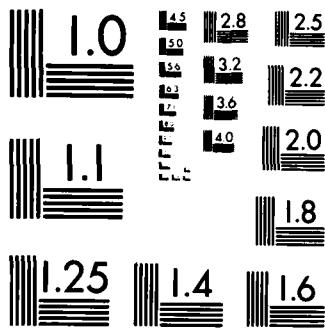


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February, 1984

Final Report

SUBNANOSECOND SHORT WAVELENGTH GENERATION USING OPTICAL FIBERS

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submitted to

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Air Force Office of Scientific Research

ABSTRACT

A one-year study of nonlinear optical effects in fibers using UV lasers investigated stimulated Raman scattering. Lines were seen due to quartz in fibers and due to organic liquids in liquid-filled capillaries. Benzene formed a waveguide, but in preliminary experiments only 1% was converted to stimulated Stokes scattering. Of particular interest was the discovery of Raman scattering of the Nitrogen laser light from the nitrogen vibrational transitions within the laser itself. Such lines have never been reported before. Of additional interest was the multi-line spectral broadening of the stimulated Raman spectra observed from quartz fibers. The reason for this broadening remains to be investigated.

Preliminary experiments were performed and apparatus designed which sets the stage for further research into the high power UV properties of fibers.

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REPORT DOCUMENTATION PAGE

1a. REPORT SECURITY CLASSIFICATION NONCLASSIFIED		1b. RESTRICTIVE MARKINGS													
2a. SECURITY CLASSIFICATION AUTHORITY		3. DISTRIBUTION/AVAILABILITY OF REPORT													
2b. DECLASSIFICATION/DOWNGRADING SCHEDULE		UNLIMITED Approved for public release; distribution unlimited.													
4. PERFORMING ORGANIZATION REPORT NUMBER(S)		5. MONITORING ORGANIZATION REPORT NUMBER(S) AFOSR-TR-85-0491													
6a. NAME OF PERFORMING ORGANIZATION Center for Laser Studies University of Southern California	6b. OFFICE SYMBOL (If applicable)	7a. NAME OF MONITORING ORGANIZATION Air Force Office of Scientific Research													
6c. ADDRESS (City, State and ZIP Code) Los Angeles CA 90089-1112		7b. ADDRESS (City, State and ZIP Code) Bolling AFB DC 20332													
8a. NAME OF FUNDING/SPONSORING ORGANIZATION AFOSR	8b. OFFICE SYMBOL (If applicable)	9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER F49620-84-C-0016													
8c. ADDRESS (City, State and ZIP Code) Directorate of Physical and Geophysical Sciences Building 410 Bolling AFB DC 20332-6448		10. SOURCE OF FUNDING NOS. <table border="1"> <tr> <td>PROGRAM ELEMENT NO. 61102F</td> <td>PROJECT NO. 2301</td> <td>TASK NO. A1</td> <td>WORK UNIT NO.</td> </tr> </table>		PROGRAM ELEMENT NO. 61102F	PROJECT NO. 2301	TASK NO. A1	WORK UNIT NO.								
PROGRAM ELEMENT NO. 61102F	PROJECT NO. 2301	TASK NO. A1	WORK UNIT NO.												
11. TITLE (Include Security Classification) SUBNANOSECOND SHORT WAVELENGTH GENERATION USING OPTICAL FIBERS															
12. PERSONNEL AUTHOR(S) Elsa Garmire															
13a. TYPE OF REPORT Final	13b. TIME COVERED FROM 1/1/84 TO 12/31/84	14. DATE OF REPORT (Yr., Mo., Day) 2/15/85	15. PAGE COUNT 15												
16. SUPPLEMENTARY NOTATION															
17. COSATI CODES <table border="1"> <tr> <th>FIELD</th> <th>GROUP</th> <th>SUB GR.</th> </tr> <tr> <td></td> <td></td> <td></td> </tr> <tr> <td></td> <td></td> <td></td> </tr> <tr> <td></td> <td></td> <td></td> </tr> </table>	FIELD	GROUP	SUB GR.										18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number) UV, optical fibers, waveguides, nonlinear optics pulses, stimulated Raman effect. 4		
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20. DISTRIBUTION AVAILABILITY OF ABSTRACT UNCLASSIFIED/UNLIMITED <input checked="" type="checkbox"/> SAME AS PPT <input type="checkbox"/> DTIC USERS <input type="checkbox"/>		21. ABSTRACT SECURITY CLASSIFICATION UNCLASSIFIED													
22a. NAME OF RESPONSIBLE INDIVIDUAL <i>C. H. Knerberg</i>		22b. TELEPHONE NUMBER (Include Area Code) <i>(202)767-4906</i>		22c. OFFICE SYMBOL <i>Y17</i>											

Final Report

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Introduction

This was a one-year program to begin research into short wavelength generation using optical fibers. Although the program was originally conceived and begun by Professor Mordechai Rothschild, he left USC in November and in the last two months Professor Elsa Garmire took over active technical direction and responsibility for the personnel. The graduate student Hisham Abad continued working on the program. The research remained on schedule, with delivery of the flashlamp-pumped dye laser operating near specifications. Initial experiments were performed on all three major aspects of the program - gas-diffused quartz fibers, liquid-core fibers and the search for phase-matched anti-Stokes generation. This report outlines the progress made in each of these areas.

Technical Discussion

1. Flash-lamp pumped dye laser

The newly purchased Candela dye laser was characterized and shown to provide 300 nsec pulses, with an energy of 2 J/pulse, using Rh-6G dye, close to the specifications of the manufacturer. The beam quality was excellent, with less than 2 mrad beam divergence. Spectral quality was not as high as desirable and depended on alignment. Typical spectra are shown in Figure [1]. Operation was either in two narrow lines 3 Å wide and separated by 7 Å or in one line of width 11 Å. This latter linewidth was ten times larger than the N₂ laser which was used in previous experiments.

The 300 nsec duration of the pulse was too long for nonlinear optics experiments in fibers. In preliminary experiments on the transmission of this power through both quartz and liquid-core fibers, the fibers blew up under the higher energy from the dye laser. For this reason it was necessary to narrow the pulse in time using a pulse-slicer, which could reduce the pulse duration by an order of magnitude. We tested the new pulse slicer and found that it produced pulses of programmable length with a minimum pulse duration of 35 nsec, close to specifications. However, we also measured a decrease in peak intensity as a function of pulse width

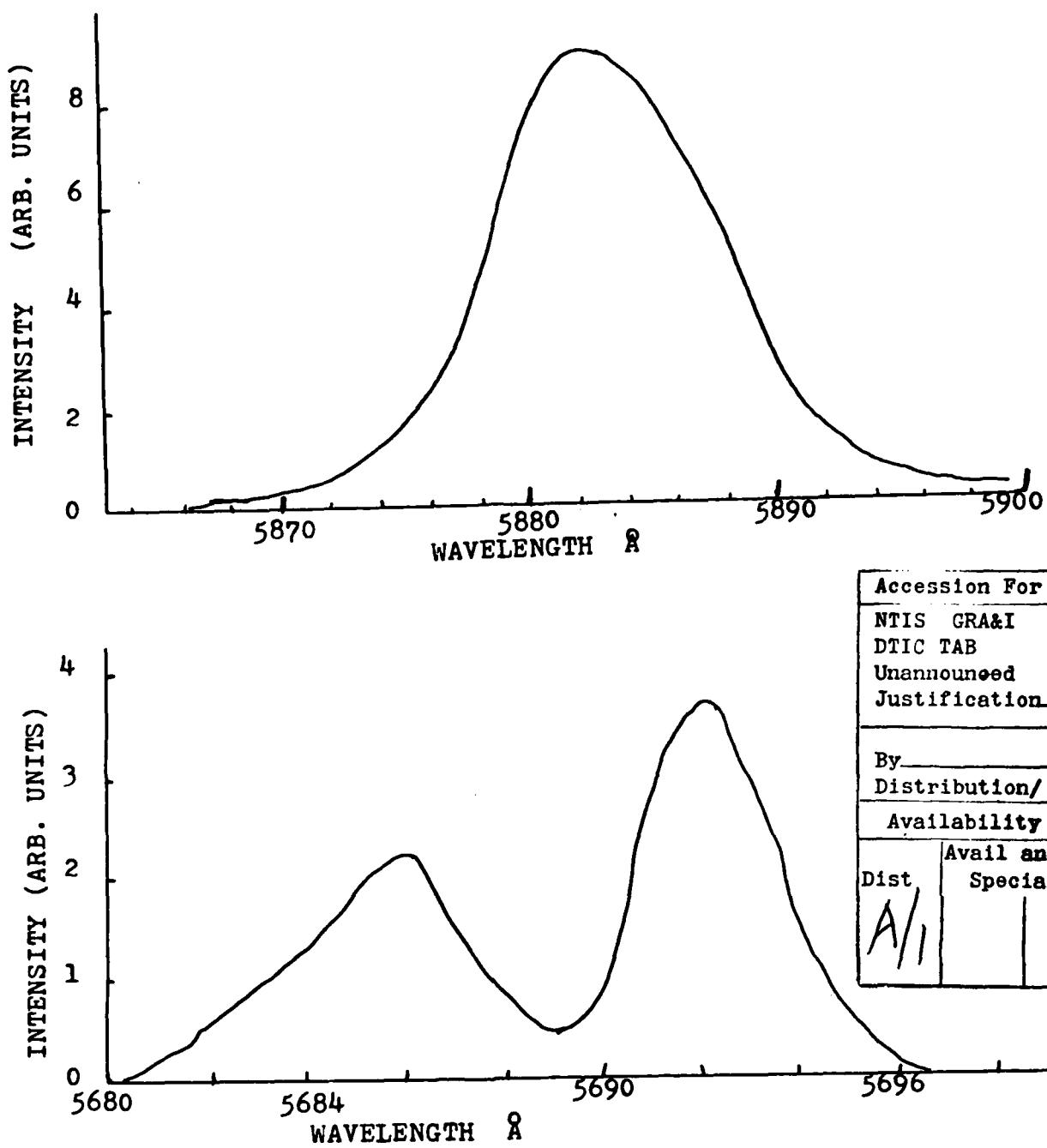


Figure 1. Spectrum of dye laser, for two different alignments.

for pulse widths less than 150 nsec. Typical data are shown in Figure [2]. This reduction in peak intensity must be taken into account in designing future experiments.

2. General experimental procedure for measurement of Raman Scattering

A pulsed laser [excimer, nitrogen or dye] was focussed by a short focal length lens (4-5 cm) into quartz fibers of 200 - 300 μm diameter. These were solid core with plastic or glass claddings or were hollow capillaries. The output was collected by a second lens and focussed onto the slit of a 0.75 m monochromator. The monochromator was scanned manually for the proper wavelength and the light was detected at the exit slit by a photomultiplier.

Difficulties in taking data arose due to the low rep rate of the lasers (1 Hz max) and the fast decay of the pulse energy with many shots. Since the pulse-to-pulse variation was great, a large number of pulses were needed for each data point. Because of the finite lifetime of the gas or dye fill of the lasers, normalization was not sufficiently accurate. This is particularly important since the nonlinear effects depend on intensity. This laser constraint indicates that in the future experiments would best be performed on a single-pulse basis, such as by using film or an OMA. The data in this report, however, were taken as described above. This is the procedure which was used by Rothschild and Abad to report the Stokes Raman line in quartz fibers [Optica Letters, October, 1983].

3. "Gas in Glass" experiments

The intention of these experiments was to provide Raman-active media within a quartz fiber in order to provide new Stokes and phase-matched anti-Stokes lines. At the beginning of this program Rothschild and Abad had already reported on the Stokes Raman line which occurs from the OH vibrations in quartz fibers. When the excitation is the nitrogen laser at 338 nm, this OH line occurs at 343 nm. While the stimulated Stokes line was observed, no stimulated anti-Stokes was seen in these experiments. This may have been because the stimulated Raman line in quartz was relatively weak (~10% of the pump). The spontaneous Raman line in quartz is relatively weak. By contrast, the Raman line in hydrogen is much stronger and it was proposed that by diffusing hydrogen into the fiber, we could provide a stronger Raman scatterer, enhancing the possibility of anti-Stokes output.

Before experiments were begun on diffused hydrogen, it was necessary to develop the high-pressure diffusion technique using a less dangerous gas. Thus diffusion of nitrogen was attempted first. We performed a series of experiments looking for the Raman line from diffused nitrogen in UV transmitting quartz fibers. A 35

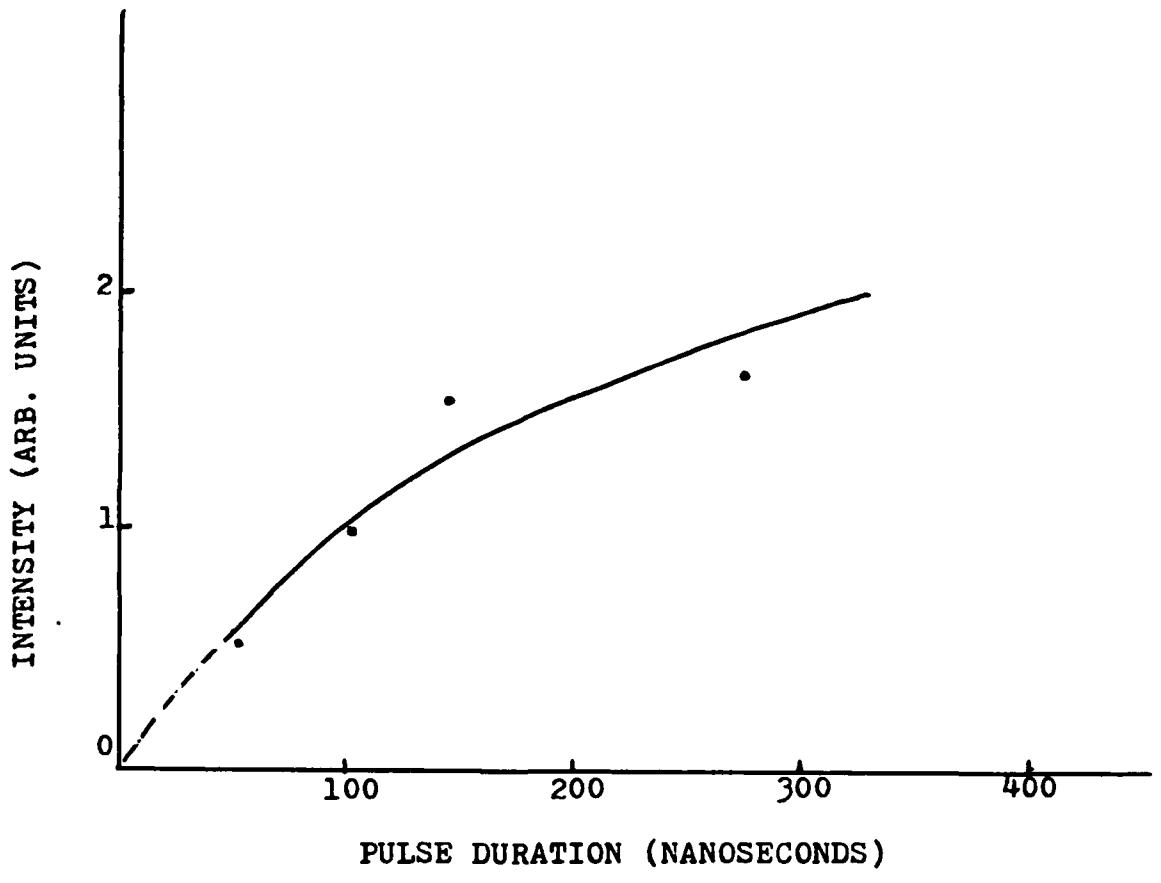


Figure 2. Intensity of pulse after Pockels' cell "pulse-slicer" as a function of pulse length.

meter long, 300 um diameter fiber was inserted in a regular nitrogen bottle, then it was pressurized to 1300 psi for a period of two months. The use of nitrogen was for practice before using the more dangerous hydrogen, which was the ultimate aim of the research. It was hoped that under pressure for two months the nitrogen would diffuse into the core of the fiber. That would have provided us with a new Raman medium which would shift the laser line by 2331 cm^{-1} . When excited by a nitrogen laser line at 337 nm, the first Stokes Raman line would appear at 366 nm.

After removing the fiber from the high pressure nitrogen bottle, we began experiments looking for the Raman line due to the nitrogen vibration. We observed a weak line at 367 nm, as shown in figure [3]. However, this line existed even in non-diffused fibers. Eventually we found that this line existed even when the fiber was removed from the experiment, and we concluded that this line arose from the nitrogen in the laser cavity. Thus we believe we have observed Raman scattering from the nitrogen within the laser itself. That is, the electronic nitrogen transition provides the initial laser line, and the vibrational nitrogen structure provides the Raman shift. To our knowledge this is the first observation of a Raman line emitted from a gas laser. It was therefore not possible to separately identify any contribution from nitrogen in the fiber and the attempt to diffuse nitrogen into fibers was stopped.

The preliminary experiments in nitrogen allowed us to test a pressure system which can be used *in situ* to study the Raman line in hydrogen by adapting a pressure cylinder. The design for the system is shown in Figure [4]. In particular, a length of fiber is inserted and epoxied into a plug which blocks the inlet and outlet of the pressure vessel. This ensures that the full pressure can be applied safely.

4. Liquid Core Waveguides

Using the nitrogen laser at 367 nm, we studied the transmission of a series of hollow waveguides of diameter 200 and 300 um, filled with various liquids. We built a liquid waveguide from both 300 um and 200 um hollow quartz capillary tubes surrounded by a larger glass tube for protection, and with two liquid cells at each end, as shown in Figure [5]. The purpose was to look for stimulated Raman scattering caused by organic liquids that formed the core of this waveguide, with the intent of ultimately generating phase-matched anti-Stokes radiation. The length of the guide was 1 meter long and designed to increase the interaction length for the nonlinear process over that of a focal spot, particularly when using a UV laser with a poor beam divergence.

Several problems arose. We had to find a suitable liquid that transmits in the UV, which has a large Raman cross-section and with a higher refractive index than that of quartz. The nitrogen laser

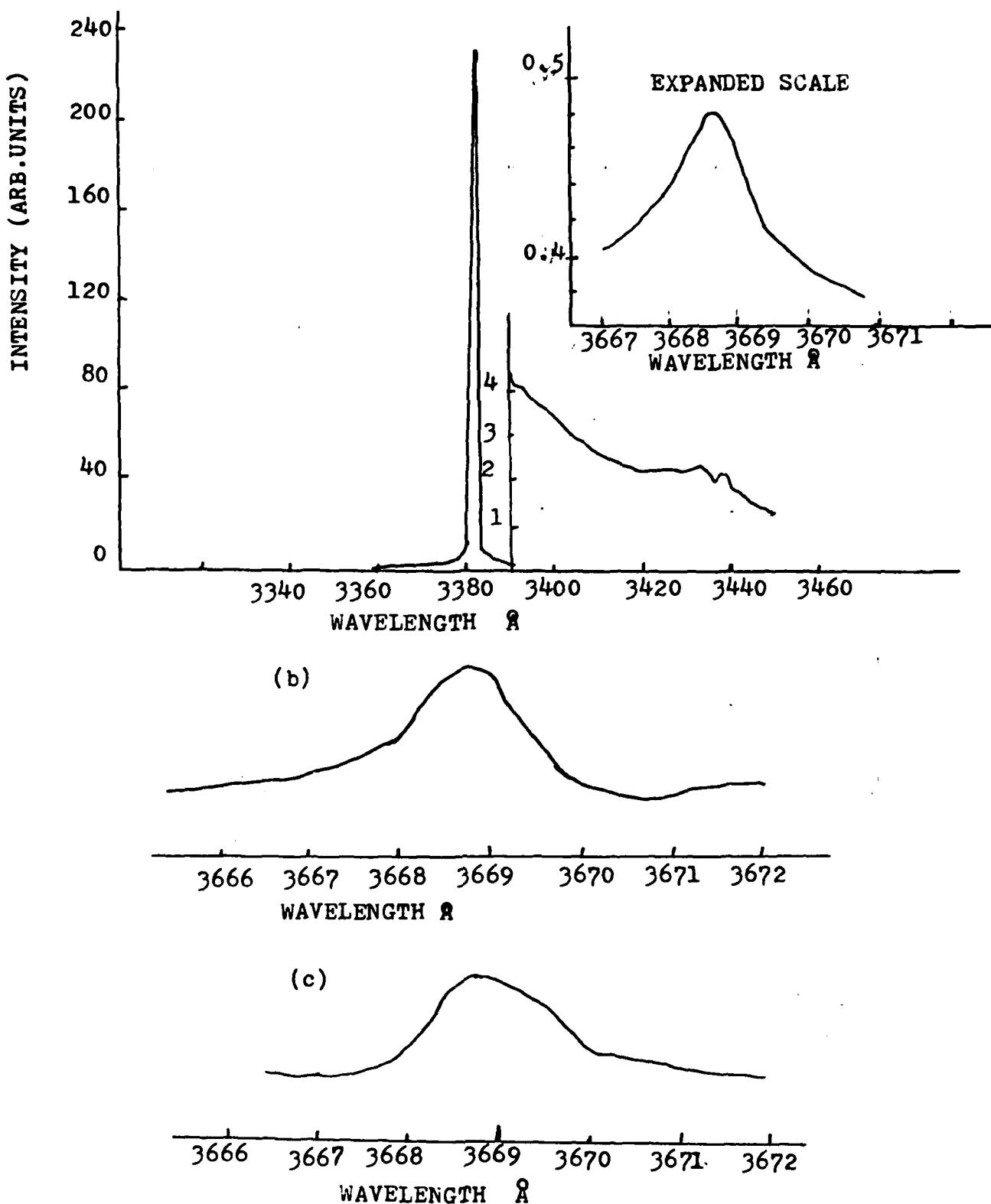


Figure 3. Spectra taken for "gas in glass" experiment. a) Spectra of nitrogen-diffused fiber. b) Spectrum of clean fiber, around raman line. c) Spectrum of N_2 laser, around Raman line.

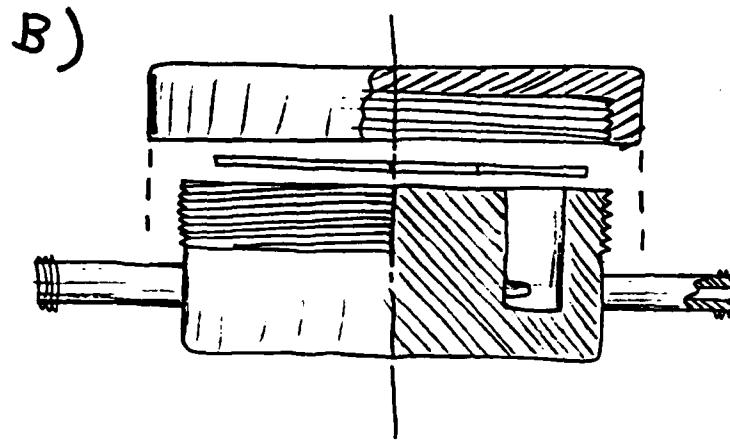
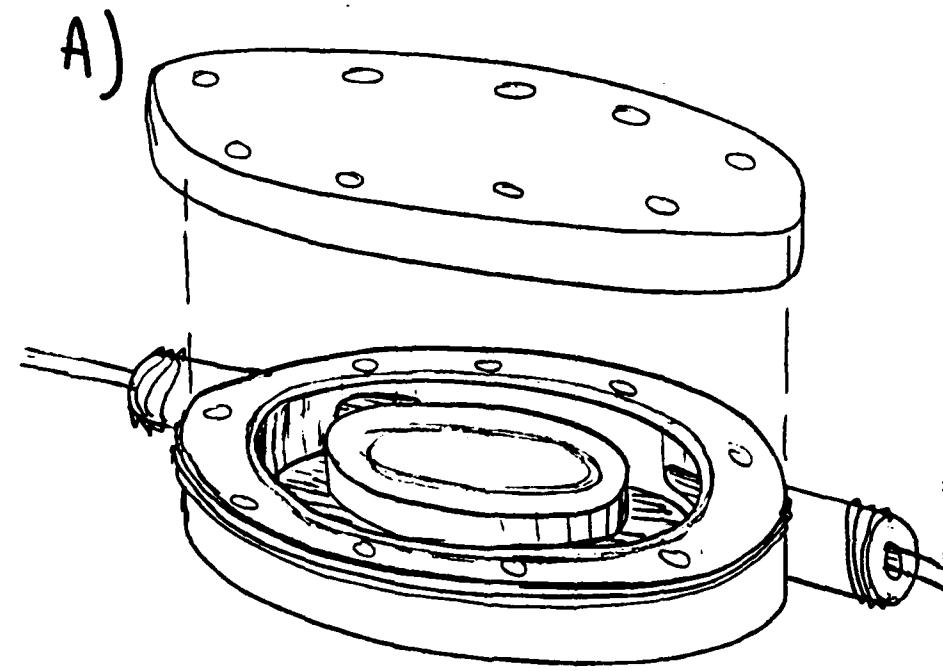


Figure . Design of high-pressure chamber of diffusing hydrogen into silica fibers. a) Opened view. Top bolts down with "O" ring seal. b) Cross-section showing threads for added protection.

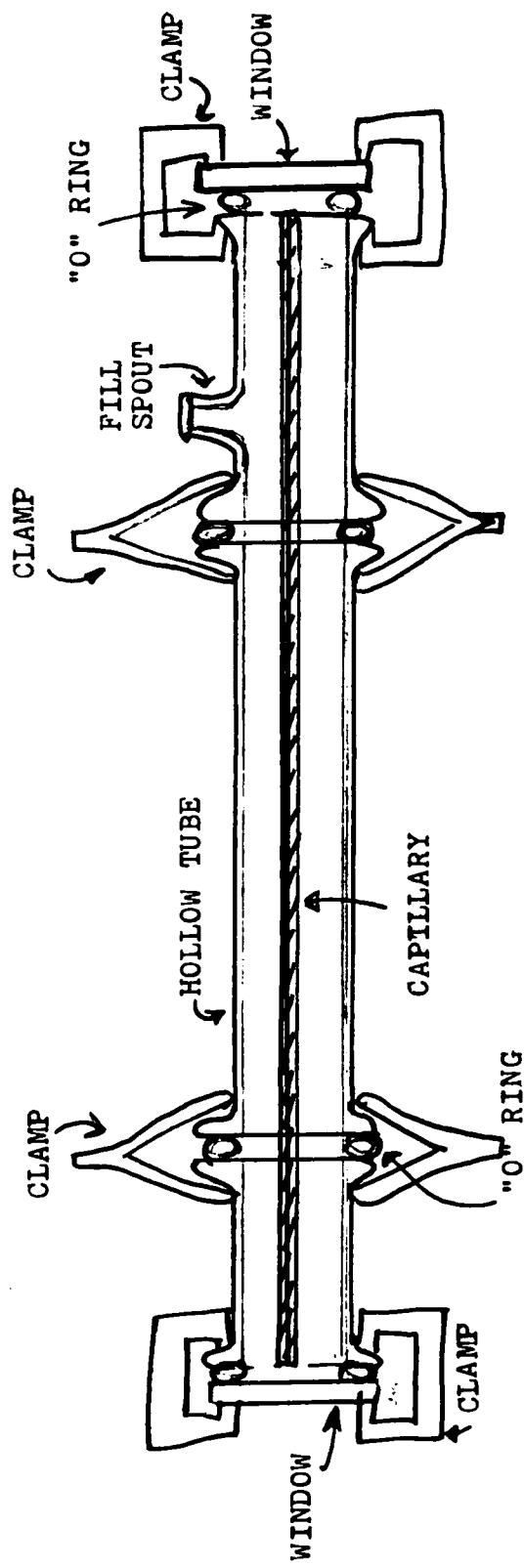


Figure 5. Liquid cell for liquid-filled capillary measurements.

was used as the pump source. Preliminary experiments were made looking for Raman scattering from cyclohexane, acetone, toluene and benzene. In general the Raman lines observed were very weak. This was either due to the absorption of these liquids in the UV or due to the lower index of the liquids than of the quartz capillary, causing anti-guiding. It was apparent the more careful measurements to determine guiding were necessary.

Benzene was investigated as the most likely waveguide for a liquid-filled capillary. Figure 6 shows the refractive index of fused quartz as a function of wavelength and also indicates the refractive indices of the four organic liquids studied. It can be seen that, at least in the visible, only benzene and toluene can form waveguides. Assuming that the relative ratio of refractive indices remains even as shorter wavelengths, it can be seen that benzene should have the highest dielectric discontinuity. We therefore studied benzene-filled capillaries. We first launched the laser into an empty capillary and measured the output. We then filled the capillary with benzene and measured the output. We found the signal as high in the presence of benzene, confirming the existence of a waveguide. Thus Benzene was found to be the most suitable to look for Stokes with the least loss. We looked at the lines shifted by 992 cm^{-1} and 3062 cm^{-1} . Again only very weak lines were observed at wavelengths. Typical results are summarized in Figure [7]. The 992 cm^{-1} and 3062 cm^{-1} shifted lines had roughly equal intensities, about 1% of the incident light. As a check, the signal without any liquid in the capillary did not show the Raman lines, as indicated in one of the enlarged inserts.

We thought that the reason for the weak Raman lines was that the nitrogen laser did not focus well because of its bad beam quality. An unstable resonator was built in order to improve the brightness and we tried the experiments again. This, however, led to a new problem. Bubbles started to appear at the input end of the capillary and that reduced the transmission of the waveguide to a minimum. Experiments were done to determine how to get rid of the bubbles. We flowed the benzene through the capillary but that did not solve the problem. We also tried to use filters to purify the liquid and that did not help either. We decided that the shorter pulses using the doubled dye laser would be better for this experiment, and we stopped working with the N₂ laser source.

5. Four-photon Generation of Anti-Stokes

Preliminary experiments searched for non-linear four-photon anti-stokes generation from stimulated stokes in quartz fibers, using the nitrogen laser. In order to reduce the phonon noise in the quartz fiber we thought that by cooling the fiber we could remove the spontaneous anti-stokes lines of the 337 nm fundamental. A 35 meters long, 300 μm fiber was cooled at liquid nitrogen, dry ice and at 0° C. No stimulated anti-Stokes and only weak Stokes

REFRACTIVE INDEX OF FUSED QUARTZ VS. WAVELENGTH

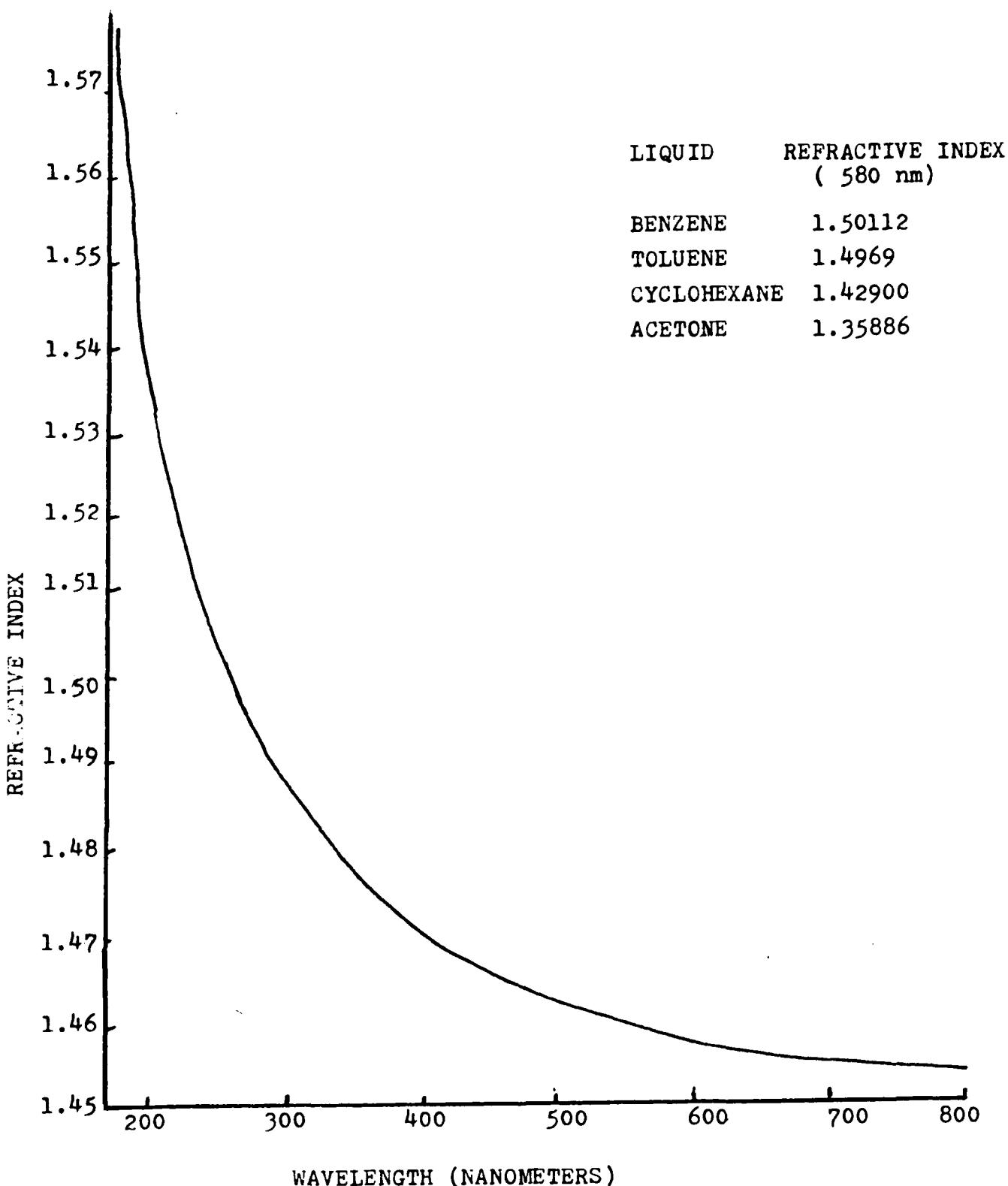
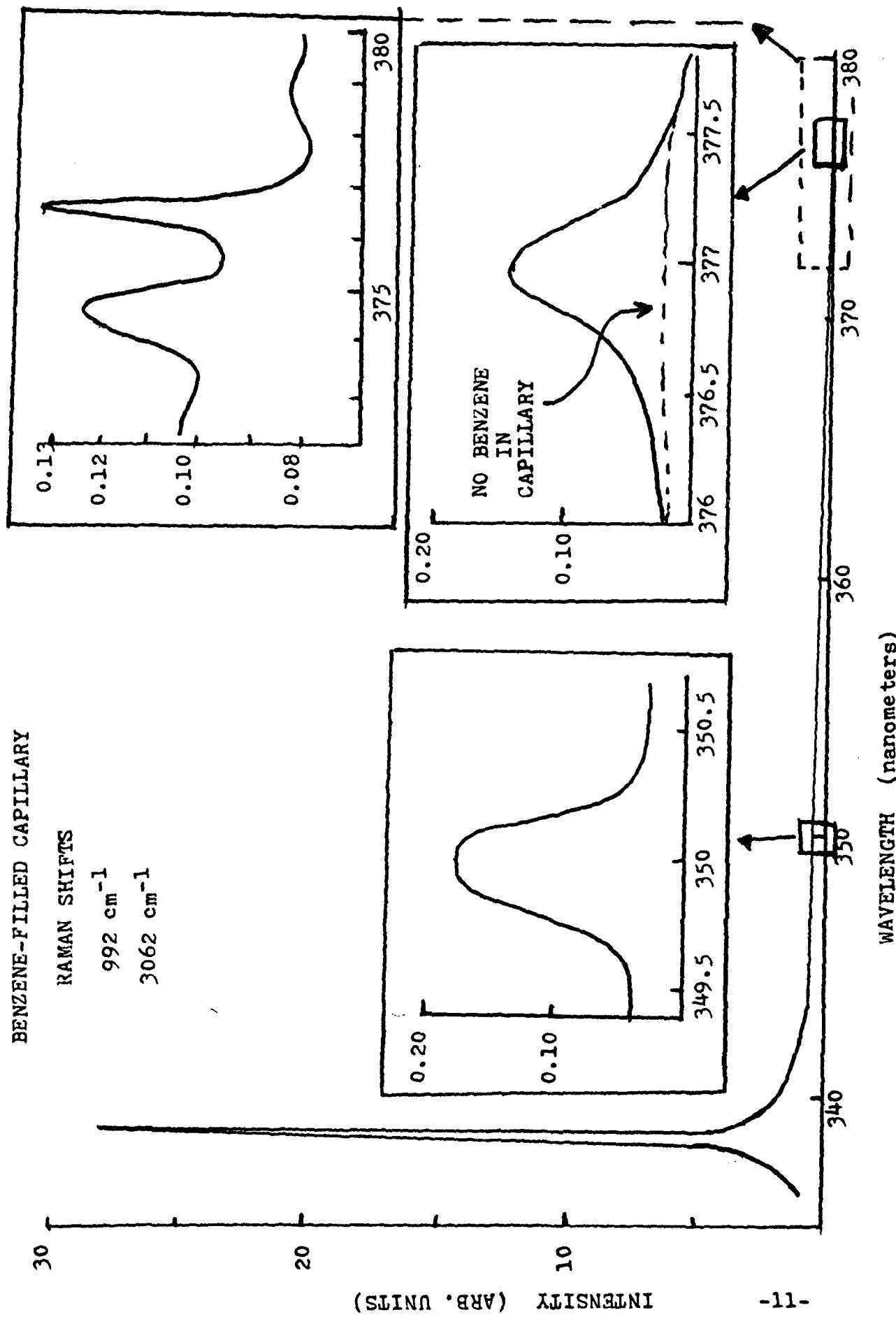


Figure 6.

Figure 7.

BENZENE-FILLED CAPILLARY

RAMAN SHIFTS
 992 cm^{-1}
 3062 cm^{-1}



lines were seen. In fact, it was found that the transmission of these plastic-clad quartz fibers dropped dramatically with temperature. This loss was reversible, but at 77° K the transmission was only 10% of its room temperature value. As the temperature increased, the transmission of the fiber went back to its original value. The reason for the strong decrease in transmission at lower temperatures was presumably due to the strong dependence of the refractive index of the plastic cladding on temperature. A weak Raman Stokes line was seen at dry ice temperatures. This is shown in Figure [8]. Because of the loss introduced by cooling, the low-temperature experiments were dropped. The data taken, however, show some interesting properties of the stimulated Raman spectra in quartz fibers. The Raman lines were typically double, and separated by 5 Å. As a comparison, the spectrum of the Nitrogen laser is shown in Figure [9] and can be seen to have a full width at half maximum power of 1.54 Å. Thus the Raman doublet is real, and presumably due to the properties of fused quartz, or to the properties of the fiber. Further study is required to understand the origin of this doublet.

Generation of anti-Stokes requires phase-matching, which cannot be calculated because there is insufficient data on the UV refractive properties of liquids. Thus we began an experiment to measure the refractive index of these materials by measuring the minimum deviation angle in a prism. This was carried out by an undergraduate as a research program. Difficulties in achieving reproducible results delayed this experiment, and it has not been completed.

As an alternative, stimulated Raman scattering in gas-filled guided wave devices was considered. Phase-matching can be achieved by the addition of buffer gases. A novel scheme has been proposed and is being modelled, consisting of using an inverted population to create stimulated anti-Stokes. The inverted population will come from a hydrogen gas discharge. In order to achieve a long interaction length within a plasma discharge, whispering gallery modes will be used, as we have previously demonstrated with a He-Ne laser [E. Garmire, M. Bass, T. McMahon, IEEE J. Quantum Electronics, January, 1980].

6. Operation of Excimer Laser

The nitrogen laser used in preliminary experiments was replaced by a XeF excimer laser in order to obtain lower beam divergence and more power. The output was measured as 60 mJ in 15 nsec, with a beam divergence of 2×3 mrad. This laser was used to investigate Raman scattering in liquids. However, because this was a stationary-fill laser, the power dropped substantially after only a dozen shots, and we found it unsuitable for point-by-point measurements of spectra which was our original experimental technique. Use of photo-graphic film for single-shot analysis would make this

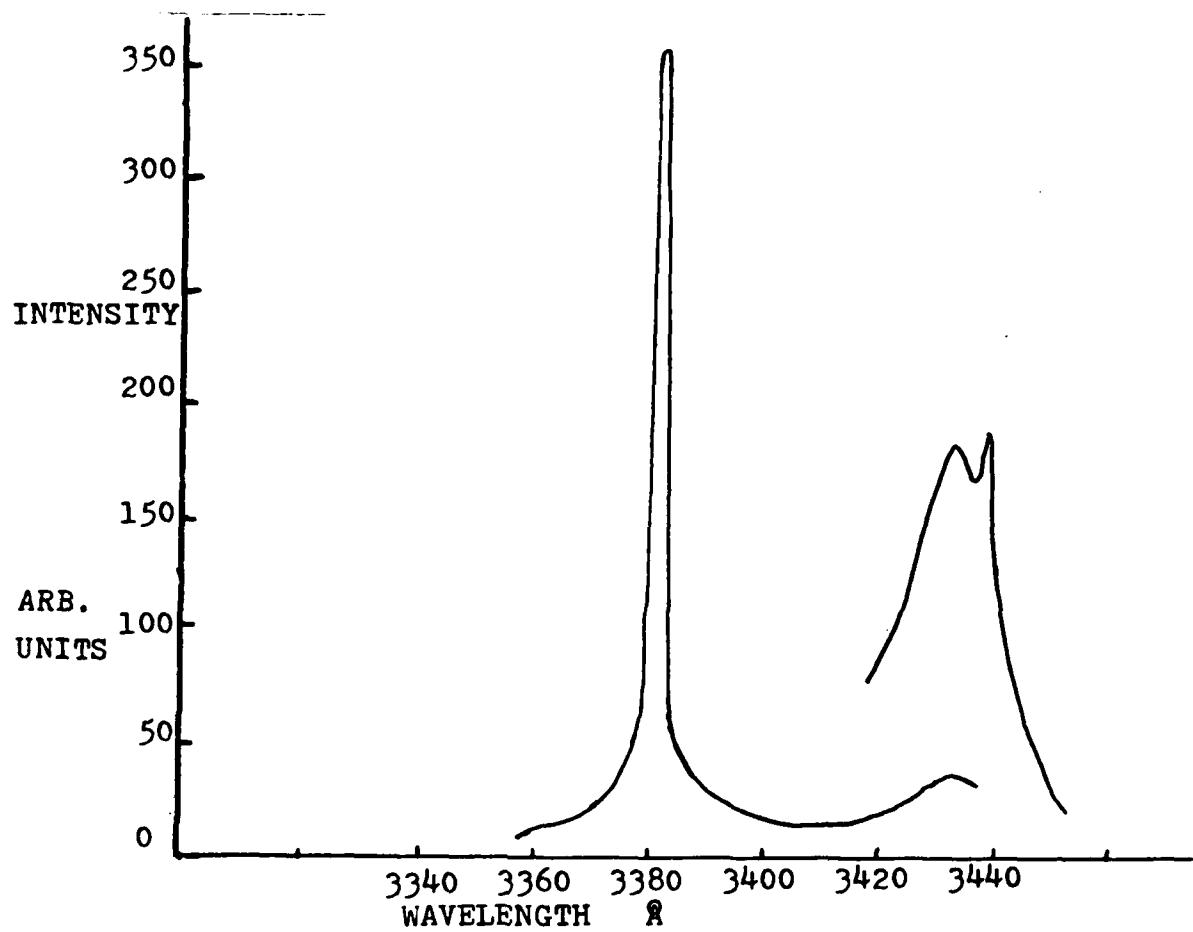


Figure 8. Stimulated Raman line in quartz fiber initiated by nitrogen laser pulse. This Raman line is due to the OH vibration in the quartz.

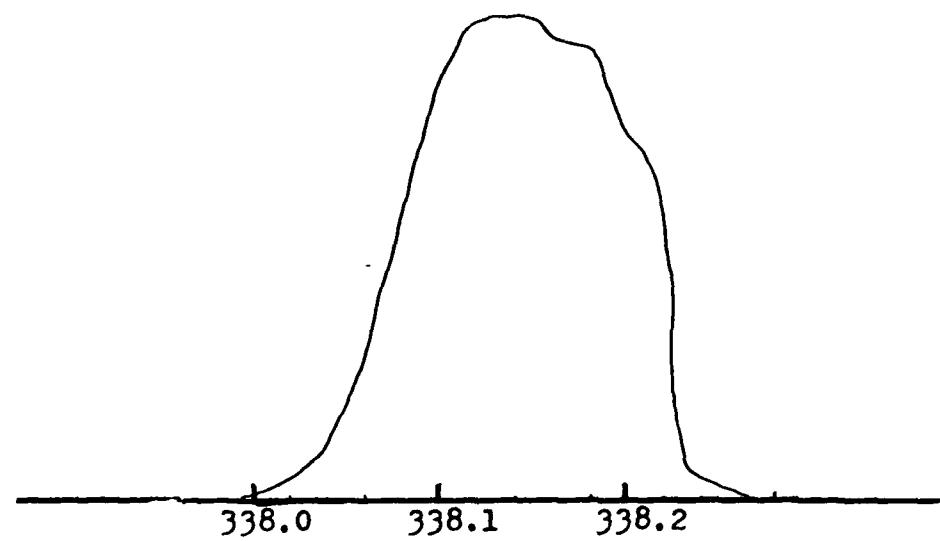


Figure 9. Spectrum of N_2 laser, spread out to indicate laser linewidth.

laser useful, and probably should have been used. Alternatively, this laser would be useful with an OMA for recording the data.

Personnel

The work was under the direction of Mordechai Rothschild until November and under the direction of Elsa Garmire after that. The student Hisham Abad worked on the project throughout the year.

Publications

Because of the preliminary nature of the work and its one-year duration, there were no publications or patent applications during the period of the contract.

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